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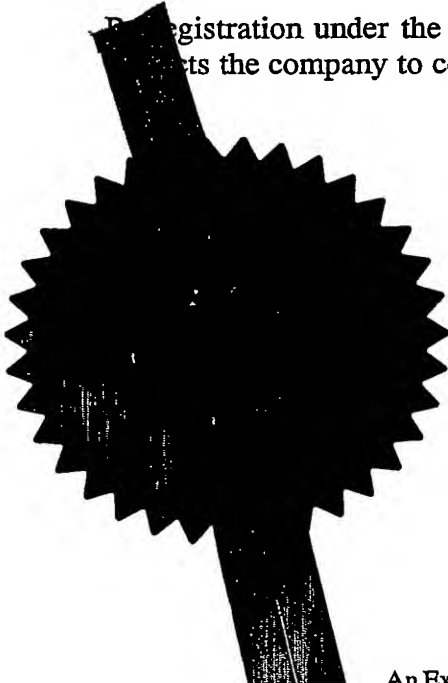
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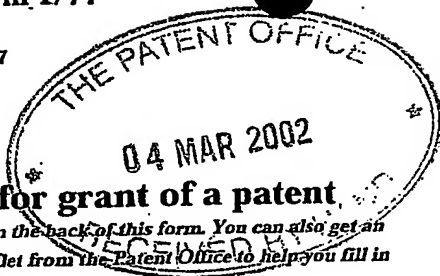
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4 MAR 2002

The Patent Office

Cardiff Road
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1. Your reference

HMJ03559GB

2. Patent application number

(The Patent Office will fill in this part)

0205021.9

3. Full name, address and postcode of the or of each applicant (underline all surnames)

OLE-BENDT RASMUSSEN
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SWITZERLAND

Patents ADP number (if you know it)

6056 337003

If the applicant is a corporate body, give the country/state of its incorporation

4. Title of the invention

Cross-Laminate of Oriented Films, Method of Manufacturing same, and Coextrusion Die suitable in the Process

5. Name of your agent (if you have one)

Gill Jennings & Every

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

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Patents ADP number (if you know it)

745002 ✓

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Country

Priority application number
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Date of filing
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Number of earlier application

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8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

NO

- a) any applicant named in part 3 is not an inventor, or
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Patents Form 1/77

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Description 26

Claim(s) 8

Abstract

Drawing(s) 4 7 Y

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Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

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Any other documents (please specify)

NO

11. For the applicant
Gill Jennings & Every

I/We request the grant of a patent on the basis of this application.

Signature

Helen Marjorie Meredith

Date

4 March 2002

12. Name and daytime telephone number of person to contact in the United Kingdom

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Cross-Laminates of Oriented Films, Method of Manufacturing same, and Coextrusion Die suitable in the Process.

Cross-laminates of oriented films from synthetic polymer materials have been commercially produced since 1968, then mainly as described in the inventor's patent GBP, 792,976 of May 23, 1955.

To the inventor's knowledge the total annual worldwide production today amounts to about 30.000 tons. The cross-laminate is used in particular as industrial bags, cover-sheet, tarpaulins, pond-liners and similar products. The polymer materials used for these cross-laminates have mainly been and are mainly polyethylene and polypropylene of different types, often modified by blending, and the old and present manufacturing processes comprise the steps of extruding a tube which by the draw-down mainly is oriented in its longitudinal direction, helically cutting this tube to a web with its main direction of orientation on bias, and continuously laminating two or more such webs with their main directions of orientation criss-crossing. There can also be included in the laminate a film which mainly is oriented in its longitudinal direction.

In the first commercialized technology based on these principles, the extruded tubular film which mainly is meltoriented in its longitudinal direction is further cold stretched in this direction prior to the helical cutting. In a later commercialized technology, disclosed e.g. in U.S. 4.039.364 (Rasmussen) each tubular film is coextruded, having a layer which mainly contributes to the tensile strength in the laminate (hereinafter "the main layer") and

at least one surface layer (hereinafter "the first bonding layer") adapted to help in the bonding-together of the films, which at least partly takes place by pressure and heat.

There is also coextruded special layers on the films which become exterior in the laminate. These special layers are adapted to modify the surface properties of the laminate, especially for improved heatsealing. In this later technology the helical cutting takes place in direct succession to the coextrusion without any coldstretching between, but in a separate production line. However, further stretching is carried out when the films have been brought together in a sandwich arrangement, bonded or not yet bonded to form a laminate. They then become biaxially stretched at a relatively low temperature.

The transverse component of this biaxial stretching takes place between grooved rollers.

In U.S. patents 5.028.289 (Rasmussen) and 5.626.944 (Rasmussen) this stretching between grooved rollers has been further developed.

Practical ways of carrying out the helical cutting are disclosed in U.S. 5.248.366 (Rasmussen). This patent also mentions an alternative, namely that the tubular film can be supplied with a helically extending melt orientation while it is drawn off from the coextrusion die, established by a relative rotation between the exit and the die, and subsequently the cutting may be straightly longitudinal or otherwise be under an angle to the main direction of

orientation. It may even be adjusted to produce a web in which the main direction of the melt orientation will become perpendicular to the longitudinal direction of the web.

For the sake of completeness it should also be mentioned that, in very early patents, there is also disclosed the possibility that longitudinally oriented polymer film material can be discontinuously cross-webbed and bonded in a press.

In a process which is entirely different from that described above, cross-laminates of a very stiff character are made for use in special, advanced products. They consist of polymers which in molten or part-molten state are fluid crystals, and which become oriented and cross-laminated already within the extrusion die by means of counter-rotating dieparts. However, this type of process and product is not a subject of the present invention.

Going back to the other type of cross-laminates, which more has the character of commodities or technical products, they are especially characterised by high puncture strength and high tear propagation resistance. The heatseal strength in a shear-type seal is adequate when a suitable lower melting polymer has been chosen for the surface layers of the laminate, while very special precautions must be taken if good shock-heat-seal strength is requested in peel-type heat-seals, as usually needed for industrial bags supplied with such heatseals. These precautions are disclosed in the inventor's patent publications U.S. 5.205.650 and WO 93/23434.

As mentioned above the cross-laminates can exhibit a particular high tear propagation resistance, however this is under the condition of a generally low bonding strength. Due to the unbalanced orientation in the individual films and the criss-crossing of the main directions of the orientation, one film will have a tendency to propagate the tear in one direction and another film will tend to propagate the tear in another direction. Thereby there will be a tendency to eliminate the bonding at the location where the forces are concentrated, and if this tendency is sufficiently pronounced, the tear will "fork out" under a local delamination, and the "notch effect" of the tearing will almost be eliminated.

Hereby there will, generally speaking, be "competition" between the adhesive forces which try to withstand delamination, and the cohesive forces in each film which try to avoid a rupture or flow along any direction which is not parallel with the main direction of orientation. These adhesive forces are (still generally speaking) independent of the thickness of the films, while the said cohesive forces are mainly proportional to the film thickness, when all other parameters are unchanged. As a consequence of this "competition", "thin" crosslaminates will either exhibit a relative poor tear propagation resistance or a relatively high tendency to delamination. This is much less of a problem for crosslaminates of "thick" layers. For bags this "competition" will usually not cause any problems since filled bags are usually not subjected to delaminating

forces, which means that the bonding strength can be chosen very low, but the matter is very important for tarpaulins, cover sheet and similar products which will be subjected to repeated flexing during use, e.g. will flap in the wind. As a matter of practical experience the inventor and his licensees have found that in a tarpaulin made from a two-film crosslaminate based on combinations of LLDPE- and HMWHDPE-types, each of the films must be of a gauge of at least 45-50 grams per square meter, otherwise either the bonding strength or the tear propagation resistance will be unacceptable to the users. These experiences concern tarpaulins for "static" uses where there will not occur much flapping in the wind. For "dynamic" uses such as cover over trucks or goods waggons, where the tarpaulin will be subjected to a strong, repeated flapping, the needed gauge is much higher.

One objective of the present invention is to solve this problem, so that high tear propagation resistance and an adequate bonding between the films can be achieved at the same time and in a practical way, even in crosslaminates of a low gauge.

In connection with the solution of the above mentioned problem, the inventor has constructed a circular coextrusion die capable of coextruding, in a practical way, a circular array of strands on a tubular film, and this construction is also an objective of the present invention.

In GBP 1,095,479 of March 3, 1964 (assigned to Metal Containers) the inventor suggested that the problem which has been identified above, can be solved by strongly welding

the films to each other in spots or lines and weakly welding them together over the rest of the contacting surfaces (a strong bond/weak bond generally being better than a strong bond/no bond). This enables the tear to "fork-out" as described above in the weak-bond areas, while an overall delamination is prevented by the strong-bond spots or lines.

For the strong welding, the patent suggests heating, ultrasonic welding, application of a solvent (preferably hot vapours) to dissolve a thin surface layer, or using quickly polymerizing monomers acting as strong binders. For the weak welding the patent suggests (using polyethylene crosslinkates as an example) to apply a gel of low molecular weight polyethylene or paraffin wax, which has been dissolved e.g. in toluene or xylene by heating and has formed gel by cooling. A thin layer of this gel including the solvent is selectively applied by printing technique before the strong welding is carried out by blowing vapours of toluene or xylene towards the film surfaces while they become united between rollers.

Alternatively there is added minor amounts of a slip agent to toluene or xylene, and this "contaminated" solvent is used in similar manner as the gel.

The Danish Patent Application 1017/67 (E.I. du Pont de Nemours and Company, Wilmington) published on February 24, 1967 claims crosslinkates of films bonded in spots or lines (which may be two arrays of lines forming a net pattern) while the rest of the contact area is (quoting the main claim) "practically not bonded". Three methods of carrying

out the bonding in spots or lines are disclosed.

One consists in applying a caoutchouc-like binder in the desired pattern. This application is said to take place by wellknown methods, but it is not further explained.

A second method consists in treating the selected areas of a surface on one of the films with chlorine, followed by lamination under pressure at an elevated temperature below the melting point of the film material.

A third method - which is described as being the preferable - is carried out by treating the selected areas of a film surface with a corona discharge, followed by lamination under pressure at an elevated temperature under the melting points of the film material. In this case a rollerformed electrode, connected to earth, is supplied with the desired pattern (which may be a net-pattern) so that the electrical discharge only takes place in the space determined by this pattern. The matching film surface is corona treated over its entire area. It is indicated that this treatment requires an effect of 20 Watt per cm width if the velocity is 0,5 meter per minute.

In the above mentioned later patent U.S. 4.039.364

(Rasmussen) in which there is coextruded a surface layer on each oriented film ("the first bonding layer") to enhance and control the bonding, a strong bond/weak bond adhesion system is established by using different lamination temperatures at the different locations of the laminate. Thus in example 1, there is first under use of coextrusion

and helical cutting made 3 films with different directions of melt orientation and surface layers of EVA as a help for the lamination (in the foregoing called "the first lamination layer"). There is established a weak bonding simultaneous with transverse orientation, by taking a sandwich of the 3 differently oriented films 7 times through a set of intermeshing grooved rollers. The pitch of these rollers is 1,5 mm of which the width of the groove amounts to 1,0 mm and the width of the circular "tooth" to 0,5 mm. Between each passage through grooved rollers, the pleats formed in the film sandwich are straightened out.

These stretching steps take place at 20°C but still produce some bonding (peel strength 10 grams per ^{cm} square metre) due to the intimate contact between the films and the effect of stretching them together. After the 7 passages at 20°C the film is passed once through a similar set of grooved rollers with the same dimensions and intermeshing, but heated to 120°C, whereby there is formed lines of strong bonding. Finally the laminate is longitudinally oriented.

In EP O 099 222 (Marcer et al) of 04/07/83, orientation and crosslamination in a spotwelded pattern is carried out as a unitary process in and immediately following a circular die with two counter rotating dieparts. Each of these dieparts produces a film supplied with an array of ribs, arranged so that the two arrays face each other. Due to the counter rotation, the melt orientation and the array of ribs become righthanded in and on one of the ribbed film, and lefthanded

in and on the other one. The two arrays of ribs are brought to meet each other at or immediately after the die exit, and bonding takes place only in the spots where the ribs intersect each other. The ribs keep the two spotwelded films spaced apart from each other also in the final product. Meltorientation with crisscrossing orientation takes place while the polymer material flows through the two counter rotating parts and by the blowing and longitudinal drawdown when the laminate has left the exit of the die. There is no subsequent orientation process carried out. The process is not a coextrusion process. The films and the ribs consist of the same polymer material and come from the same extruder.

To the knowledge of the inventor, none of the above mentioned methods of making strong bond/weak bond or strong bond/no bond adhesion patterns in crosslaminates has ever been used for commercial production although the principal, big advantage of such bonding systems in crosslaminates has been recognized for about 40 years. However, each of the proposed methods have serious drawbacks. The methods which make use of organic solvents for polyolefins, especially in vapour form, are connected with health hazards unless very expensive machinery is used, not least because it is difficult to avoid traces of the solvent to remain in the final product.

The proposed coronatreatment in a pattern, followed by lamination under pressure and heat but below the melting point of the polymer material, is applicable only if the

production capacity is very low. In commercial production of crosslaminates for commodity uses, such as e.g. tarpaulins and cover sheets, the lamination velocity must be about 60 metres per minute or more and the width about 150 cm or more. Using the above mentioned information about power consumption, the 60 m per min. and 150 cm will require 900 kilowatt, which of course is not practically possible.

Neither is treatment with chlorine in a pattern a process suited for industrial production on a big scale.

The use of binders, applied by printing technique from a dispersion or solution, requires a preceding very strong surface treatment, when the polymer material is polyethylene or polypropylene, normally a very strong treatment by corona, and therefore this method is not economical either.

A strong bond/weak bond or strong bond/no bond pattern achieved by different temperatures will inevitably create differential shrinkage if the pattern is a line pattern (including a net pattern), and this makes the crosslaminates look untidy. Differential shrinkage can be avoided if the areas of strong bonding are small dots, but in this case the product gets a dotted look which is felt unpleasant. (The inventor have had samples of such "dotted" crosslaminates tested by several customers of his licensees, and the reaction has been negative due to the appearance).

To this comes that the apparatus needed for adequate heating in a spot pattern to a temperature not too high and not too low is relatively complicated, when the velocity is high, that means the laminate must follow hot spots on a heater

over a long passage without any displacement of the laminate taking place in spite of its tendency to shrinkage.

In the unitary crosslamination process with counter rotating disparts it is, from the point of view of strength, a drawback that filmforming and molecular orientation are so closely coupled together. This makes it virtually impossible to "tailormake" the properties for different purposes. Furthermore the inventor has found that a crosslaminate which is entirely unbonded except in spots, exhibits a relatively low yield point and high tendency to creep in the middle direction between the main directions of orientation in the two laminated films.

The method according to the present invention concerns the manufacture of a crosslaminate comprising at least two neighbour films (A and B) each having a main layer, which is selected for high tensile strength, A and B being bonded to each other in a strong bond/weak bond or a strong bond/no bond pattern under use of coextruded bonding layers (which above have been referred to as "the first bonding layers") from a polymer material different from the material from which the main layer is made, and the method is characterised in that in the coextrusion process each of the said first bonding layers is made a discontinuous layer consisting of an array of strands, and in the lamination the array of strands on A are arranged to cross the array of strands on B, and while A and B are in contact with each other heat is applied generally evenly all over A and B and is adapted to make the strands on A strongly bond to the

strands on B in the spots where they intersect the latter but make a weaker bonding or avoid bonding over the parts of the contacting surfaces, which are devoid of any first bonding layer.

The resultant product is defined in claim 1.

In the coextrusion process, A and/or B are preferably also supplied with a continuous surface layer (hereinafter the second bonding layer) which is coextruded on the main layer under the array of strands, whereby the composition of the second bonding layer is different from the compositions of the main layer and of the first bonding layer, and is selected to produce, during the lamination, bonding also at locations which are devoid of any first bonding layer, but a bonding of lower strength than the bonding in the spots.

The method according to the present invention and the product made by this method do not suffer from any of the draw-backs mentioned above. The method is very suited for commercial manufacture of commodity products; there is of course not any health hazards involved; the extra costs compared to the lamination with an even bonding all over is negligible; extrusion, stretching and laminating are in essence separate process steps so that each can be optimised for the desired end use, the look of the product needs not suffer from the effects of differential shrinkage or a "dotted" appearance, and commonly used machinery for manufacture of crosslaminates can be applied, only with inexpensive additions to existing coextrusion lines (as it shall be explained below).

A very important advantage of the method and the resulting crosslaminates is that the pattern of lamination comprises not only two, but three elements, provided the coextrusion apparatus comprises the means for extruding the mentioned "second bonding layer" (even when the coextrusion apparatus with said means are not always used to extrude this layer).

These three elements in the pattern of lamination are:

- a) each spot where two strands of "first bonding layers" intersect each other,
 - b) each little area in which both contacting surface parts are devoid of any "first bonding layer", and
 - c) the areas in which there is "first bonding layer" on one of the two contacting surfaces and no "first bonding layer" on the other one.
- a) and c) together form the net pattern.

By adapting the bonding strength a), b) and c) differently for different uses, but using the same machinery, this bonding system can be very helpful for "taylor-making" of the crosslaminates.

Thus, as an example, there are certain tarpaulinlike uses where the gauge should be brought down as much as possible for cost reasons, tear propagation strength and ultimate tensile strength is of primary importance, yield tension is of relatively low importance, the aesthetics unimportant, but the resistance to delamination must be very high due to flepping in the wind. In that case a strong bond/no bond pattern is preferable, and the coextrusion of "second

bonding layer" is omitted. (The main component may be applied not only from its own extruder and through its own channel system, but also from the extruder and through the channel system which otherwise is used for the "second bonding layer"). The bonding is established as a strong welding in the spots (a) where the strands intersect each other.

In other cases there can be a need to establish a strong bonding not only in the spots (a) but also in the areas (c), while there should be some bonding, but a pronouncedly weak bonding in the areas (b).

This can also be achieved by a suitable choice of polymer materials for the "first" and "second" bonding layers (in this case the "second bonding layer" must of course be applied). The combination of strong bonding in a net pattern, and some but weak bonding over the rest of the area is a very interesting pattern of lamination, usually better than strong spotwelding combined with weak bonding over the rest. In the last mentioned case an accidentally started delamination will generally propagate over a wide area if the crosslaminate is repeatedly flexed, e.g. when it is flapping in the wind. The films will still be held together where they are spotwelded, but the rest will become unbonded and thereby loose the esthetics and to some extent the yield strength and creep resistance.

Unlike this, a weak bonding surrounded by strong bonding in net pattern, will not be allowed to propagate an accidental delamination in similar manner.

However, it should be mentioned that there also exist

applications in which the best combination is:

(a): strong welding

(b): weak bonding

(c): also weak bonding, but stronger than (b).

Preferably each of the two films A and B should mainly consist of polyethylene or polypropylene, e.g. the main layer can advantageously consist of HDPE or LLDPE or a blend of the two, the second bonding layer mainly of LLDPE but with admixture of 5-25% of a copolymer of ethylene having a melting point or a melting range within the temperature interval 50-80 degr. C, while the strands mainly can consist of a copolymer of ethylene having a melting point or a melting range within the temperature interval 50-100 degr. C or a blend of such copolymer and LLDPE containing at least 25% of the said copolymer.

The distance from middle to middle of neighbour strands in each array should normally be between 2 mm and 8 cm, preferably no higher than 4 cm, and more preferably no higher than 2 cm.

The bonding strength in the spots (a) as measured by peeling should normally be at least 40 gram per cm and the bonding strength in areas (b) similarly determined at the highest 75%, but preferably no more than 50% of the bonding strength in (a).

Unlike the crosslaminates made with counterrotating dieparts and comprising criss-crossing arrays of ribs (mentioned above in the section re. known art) the thickness increase in each of the films A and B at the locations where the strands are coextruded, should normally amount to at the highest 30% seen relative to the immediate surrounding, preferably at the highest 20% and still more preferably no more than 10%.

The coextrusion of at least one of the films A or B is preferably carried out by means of a circular coextrusion die, to form and draw-down a tubular film. Hereby the draw-down is adapted to produce a significant uniaxial or unbalanced biaxial meltorientation with the main direction of orientation and the direction of the array of strands either extending along the longitudinal direction of the film or, by means of a relative rotation between the exit of the die and means to take up the film after the extrusion is brought to extend helically along the tubular film.

Subsequently the film is cut open under an angle to the main direction of orientation and to the direction of the array. However, it is also within the scope of the invention to extrude both films A and B from a flat die and cross-web the films under use of a hot press, preferably after longitudinal cold-stretching of both.

The crosslaminates of the present invention is not necessarily limited to the two films A and B, but can comprise 3 or more layers. Thus as an advantageous construction, it may comprise two pairs of array-bonded

films A and B, especially in the arrangement A-B-A in which B has on both of its surfaces an array of strands, i.e. a "first bonding layer" and preferably also a "second bonding layer".

In another suitable arrangement comprising more than two films A and B there is additionally applied at least one more film in the lamination. Said film is also produced by coextrusion and is thereby provided with a surface layer of a composition adapted to control its bonding in the laminate. This composition and the lamination conditions are chosen such that the strength of this bonding becomes higher than the bonding strength between A and B at the locations which are devoid of the coextruded strands. Thus a delamination of the additional film is counteracted.

The surfaces of the laminate should preferably each consist of a layer adapted to improve the heatsealing properties of the laminate and/or increase its frictional properties. Such layers are coextruded in the films used as outer films in the laminate.

Normally the molecular orientation in each film A and B, which may be uniaxial or unbalanced biaxial orientation, should not be limited to that achieved in connection with the extrusion. There may be carried out a further longitudinal stretching prior to the helical cutting.

Alternatively or supplementary, the films may be further oriented by stretching in the longitudinal and/or in the transverse direction following the bringing-together of the films in a sandwich arrangement for lamination. This may take place after the heatbonding of said sandwich

arrangement to a laminate.

In the foregoing it was stated that commonly used machinery for manufacture of crosslaminates can be applied, only with inexpensive additions to existing coextrusion lines. This concerns the coextrusion of the array of strands, the discontinuous "first bonding layer". The inventor has found that this can be done by adding special but rather simple and cheap machineparts at the exit of almost any existing design of coextrusion dies. Of course there is also need for one more extruder, but the strands will normally occupy only about 2-5% of each of the extruded films, and therefore this can be a small and inexpensive extruder.

The extrusion die according to the invention is a circular extrusion die comprising a distribution part in which at least a first molten polymer material can be formed into a generally even circular flow, and bodily separate from this an exit part comprising a circular main channel with generally cylindrical or conical walls, which channel may comprise a flat zone, to conduct said molten polymer material towards an exit orifice from which it will leave the die as a tubular film structure. The special features of the invention is that said exit part also comprises a channel system for circumferential extrusion of a circular array of narrow strands of a second molten polymer material, this channel system ending in a circular row of internal orifices in the outward generally cylindrical or conical wall of the main channel. In a preferred embodiment, the circumferential extrusion

starts at one or a few inlets to the exit part and comprises for equal dividing a labyrinthine channel system starting at each inlet, each such system comprising at least three channel-branchings.

The term "labyrinthine dividing" was introduced in US Patent 4,403,934 and means a dividing of flows in which one divides into two branches of equal length, each of these again into two of equal length etc., all branches mainly being circular and parallel to each other. This is shown in fig. 4.

In order to make a particular short distance between the internal orifices, the channels of the labyrinthine system or systems may terminate in a common circular channel having a wall common with a part of the generally cylindrical or conical wall of the main channel. The circular row of internal orifices is located in said wallpart.

This coextrusion die has been conceived with a view to the manufacture of the crosslaminate as means to achieve strong bond/weak bond or strong bond/no bond lamination patterns, and for this purpose there is normally a need for a continuous "second bonding layer". Therefore, there is preferably in addition to the means for coextruding the said first and second molten polymer materials, provided means for coextruding a circular flow of a third molten polymer material on the side of the first material which is opposite the second material. Channel arrangements for joining the flows of first and third materials are provided either in the said distribution part, or in a part between the latter and the bodily separate exit part.

In the die, the circumference of the inward wall at the exit is preferably at least 20 cm, and the distance from middle to middle of neighbour orifices in the circular row is adapted to produce, after the magnification or reduction which will happen if the walls of the main channel are generally conical, a distance from middle to middle of neighbours of the strands which is at the highest 8 cm, preferably no higher than 4 cm and more preferably no higher than 2 cm.

The die can also advantageously be applied in the production of polymer film other than crosslaminates, e.g. film with a decorative pattern of coloured stripes. Besides the above mentioned economical advantage, namely that existing die designs can be used with addition of cheap dieparts and a small extruder, it is also an advantage that the route of flow from extruder to die exit becomes shortest possible when the inlet to the die and the entire distribution system is near the exit from the die, whereby degradation of the polymer best possible can be avoided.

For the sake of completeness it should be added that the array of strands in the coextruded films A and B for the described crosslaminates, of course also can be formed from a flow which passes through the entire distribution part of a coextrusion die parallel with the other flows, but then there can be a risk of degradation since these strands as mentioned above usually only will constitute about 2-5% of each film.

The invention shall now be further described with reference to the drawings, of which:

Fig. 1 is a view on a true scale of the interphase between the two coextruded, oriented, helical cut and cross-laminated films (A and B), each normally but not always with a thin continuous layer for weak bonding and on top of this layer, a thin discontinuous layer in form of an array of strands for strong bonding, so that three different elements of bonding (a, b and c) is established generally with different bonding strength in each element.

Fig. 2 is a schematic perspective drawing of a coextrusion line for manufacture of tubular film suitable, after helical cutting, for making the cross-laminate shown in Fig. 1. The flow of polymer material for the strands comes from a very small extruder (4), and flows of three other polymer materials (for main layer, continuous bonding layer, and layer for surface of the laminate) come from the bigger extruders (5), (6) and (7). The last mentioned three materials are fed into the distribution part (8) of the coextrusion die and are here each formed into a circular flow. Joining of these flows takes place at the exit from (8) while they enter the bodily separate exit part (9). The flow from the small extruder (4) is fed directly into exit part (9) where, starting from the circumference, it becomes evenly distributed in a labyrinthine channel system as shown in Fig. 4, and applied through a circular array of internal orifices on the outside of the tubular joint flow of the other components. Fig. 3 is an axial section, shown on a scale about half of true scale, of the bodily separate exit part (9), which consists of the sub-parts (9a), (9b), (9c), (9d) and (9e). The section goes through the line x-x in fig. 4 (10) are channels in the labyrinthine system

Fig 3a is a detail from fig. 3 showing the last branch in the labyrinthine channel system and one of 64 internal orifices (11) through which the flows of strand-forming material join the tubular flow (12) of the three other

coextruded materials. This detail is drawn on a scale about six times true scale.

Fig. 4 is a perspective view of sub-part (9a), showing the lower half part of the labyrinthine channel system (10), by which one flow of the strand-forming material from extruder (4), fed through the inlet (13) stepwise is divided into 64 equal part-flows through the channels (10), each extruded separately through an internal orifice (11). The upper half part of the labyrinthine channel system, which is in sub-part (9b) is exactly symmetrical with that in fig. 4.

In fig. 1 the longitudinal direction of the cross-laminated web is shown with the arrow (1). The main direction of orientation, which may be a uniaxial orientation or an unbalanced biaxial orientation, is shown by the arrow (2) for one film, and the arrow (3) for the other film. They are each shown under an angle of about 60° to the longitudinal direction (1). This is what the inventor generally has found best for tarpaulins and cover sheet, while angles near 30° generally have been found best for cross-laminates used to make bags. The angle 45° was only found preferable in a few cases.

For each film the main direction of orientation is shown almost but not quite parallel with the array of strands in the film. If the tubular film, when leaving the extrusion die, has been drawn-down straight, and if it has not been stretched after the helical cutting, the main direction of orientation will be exactly parallel with the array, but if the tubular film has been screwed during the drawn-down, to produce a helical melt-orientation, or if it has been uniaxially or biaxially oriented after the helical cutting, as in the above-mentioned US 4,039,364 (Rasmussen), the main direction of orientation will not be quite parallel with the array.

In the spots (a) where one array crosses the other one, there is established a strong spot welding. It should normally be so strong that the laminate will rupture around these spots if delamination is tried.

If there is not coextruded a continuous bonding layer (in the claims called ("the second bonding layer") between the strands and the main layer, there will only be bonding in the spots (a), but as mentioned this will be a very strong bonding. As it has been explained in the general description, this simple bonding system is preferable in some cases. However, the coextrusion die should always comprise a channel system for "the second bonding layer" so that its use is not limited to the said cases.

Most normally there should be coextruded a "second bonding layer" in each film between "main layer" and the array of strands. In the areas marked (b) there is direct adhesive connection between the "second bonding layers" in the two films, and by the choice of material composition and laminating temperature there is established a predetermined, well controlled weak bonding here.

In the areas (c) the strands on one film are adhesively connected with "second bonding layer" on the other film. Thus the bonding strength in areas (c) gets a value somewhere between those in (a) and those in (b). There is a wide choice between these three values, and the adhesives system can so to say be "tailor made" for the intended use of the cross-laminate.

With relatively thin film it is possible to make the bonding in the (c) areas so strong that it cannot be eliminated without rupture of the material, even when the bonding in the (b) areas is made particularly weak. Hereby these areas of weak bonding are fully "encased" in areas of strong bonding. The advantages of this for certain uses are explained in the general description. In other cases, it can be preferable, still while making the bonding in the (b) areas very weak, to give the bonding in the (c) areas a suitable "in-between" value which allows delamination during tear propagation, but exerts so high resistance against this delamination, that tear energy is absorbed and rupture around the (a) spots is prevented.

Figs. 2, 3, 3a and 4 have already been sufficiently described for a principal understanding, but the following should be added:

(14) shows different rows of bores for bolts or screws to keep the parts strongly together. (15) in fig. 3 shows a shallow channel for drainage, as usual in the die construction. In fact there should be a system of channels for drainage occupying most of the area between sub-part (9a) and (9b), but for the sake of clarity only this channel is shown. With reference to fig. 3a the downstream side of the internal orifices (11) is given a shape which prevents damage of the axial flow, a damage which can occur if there had been a sharp edge on this side of the orifices.

It has been emphasised that there is an exit part (9) bodily separate from the distribution part (8). As shown (9) will normally consist of several sub-parts. However, the shown sub-parts (9c) and (9e) can be constructed as one part. The centering of sub-part (9d) is made adjustable in order to compensate for thickness variations in the extruded tubular film (15).

While the distribution of the flow from extruder (4) here is shown as a labyrinthine system, which is considered preferable, it can also be other known circumferentially fed circular distribution systems.

In fig. 4 each of the final 64 branches of the labyrinthine system ends in an internal orifice which extrudes directly into the axial tubular stream. However it is not practical to divide into more than 64 branches, and if a bigger number of strands is wanted, each final branch may open into a common ring-formed channel close to the channel (12) for the axial flow. From this ring-formed channel there can be the desired big number of openings into channel (12).

Example

The procedure is the same as in US 5,028,289 (Rasmussen) example 3 except for the following:

The coextrusion line is constructed as shown in figs. 2, 3, 3a and 4, and there are coextruded strands consisting of a metallocene (catalysed) copolymer of ethylene and octlene having melting range 50-60°C and melt flow index 1.0.

The layer which in said US patent is called "lamination layer" and here "2nd bonding layer" is a blend of 90% LLDPE and 10% of the low melting copolymer. The LLDPE has density 0.92g.ml and melt flow index 1.0. The "main layer" and the "heat seal layer" (for heat-sealing of the final cross-laminate) are the same as in the said example 3. The "main layer" forms 75%, the "heat-seal layer" 15%, the "2nd bonding layer" 8%, and the strands 2% of the film.

The angle of cutting is 57°

The temperature for lamination, stretching processes, and final heat treatment are also different, namely:

For pre-heating: 60°C

For the transverse stretching between the special grooved rollers described in the said US patent and the first longitudinal stretching process: 35°C.

For the following transverse and longitudinal stretching processes: 35°C.

For the heat treatment which effects the final bonding: 90°C.

Like in the said example 3 the gauge of the final cross-laminate for identification below called I is about 70gm⁻². This has the bonding pattern shown in fig. 1.

A similar cross-laminate (called II) but without the strands, is manufactured for comparison.

Furthermore there is manufactured a third laminate (called III) similarly to I, but without the strands and with 15% instead of the 10% metallocene copolymer in the "2nd bonding layer".

(I) exhibits the highest tear propagation resistance, (II) almost the same, and (III) a considerably lower tear-propagation resistance under shock-tearing unacceptable for tarpaulins. This property is evaluated by "hand-tearing" at a measured velocity between 5 - 7ms⁻¹ by a team of people used to such testing and knowing the requirements of customers. To the knowledge of the inventor there does not exist any standardised test for tear-propagation-resistance which come close to the practical conditions of tearing.

(I), (II) and (III) are also cut into 8cm wide bands which each are set up like a flag on a stick of diameter 25mm and cut so that it extends 25cm from the pole. It is then tested by an artificial wind of about 100 km per hr. (II) and (III) delaminated within a few minutes, while (I) withstood delamination, except at the edges, for the 2 hour period which the test lasted.

CLAIMS

1. A crosslaminatc comprising mutually bonded polymer films of which at least two neighbour films (A and B) are uniaxially oriented, or unbalanced biaxially oriented, A and B each being coextruded films and each containing a layer consisting of a polymer material selected for high tensile strength (hereinafter the main layer) and a surface layer (hereinafter the first bonding layer), consisting of a polymer material different from the material from which the main layer is made and selected to produce or enhance the bonding between A and B, whereby the main direction of orientation in A crosses the main direction of orientation in B, and in which the bonding between A and B is strong in spots or lines, while there is no bonding or only a weak bonding between A and B over the rest of the contacting surfaces, characterised in that each of the said coextruded first bonding layers is a discontinuous layer consisting of an array of coextruded thin strands, the array of strands on A crossing the array of strands on B, whereby A and B have become strongly bonded to each other in each spot where a strand on A intersects with a strand on B, while A and B are weaker bonded or not bonded over the parts of their contacting surfaces, which are devoid of any first bonding layer.

2. A crosslaminatc according to claim 1, characterised in that the thickness increase in each of said films A and B at the locations where the strands are coextruded amounts at the highest to 30% seen relative to the immediate

surrounding, preferably at the highest 20% and still more preferably no more than 10%.

3. A crosslaminata according to claims 1 or 2 characterised in that the distance from middle to middle of neighbour strands in each array is between 2 mm and 8 cm, preferably no higher than 4 cm, and more preferably no higher than 2 cm.

4. A crosslaminata according to any of the preceding claims characterised in that at least one of the said films A and B contains on its side contacting the other one, a continuous surface layer (hereinafter the second bonding layer) which is coextruded on the main layer under the array of strands, whereby the said second bonding layer consists of a polymer material different from those in the main layer and in the first bonding layer, and is selected to produce bonding also at locations which are devoid of any first bonding layer, but a bonding of lower strength than the bonding in the spots.

5. A crosslaminata according to any of the preceding claims, characterised in that the bonding strength in the spots as measured by peeling, is at least 40 gram per cm and the bonding strength in the parts of the contacting surfaces which are devoid of any first bonding layer, similarly determined is at the highest 75%, and preferably no more than 50% of the bonding strength in the spots.

6. A crosslaminata according to claim 1, characterised in

that it comprises two such pairs of array-bonded films A and B.

7. A crosslaminata according to claim 6, characterised in that one film is common for two such sets, this film having an array of the said strands on both of its surfaces.

8. A crosslaminata according to any of the preceding claims, characterised in that each of the said two films A and B mainly consists of polyethylene or polypropylene.

9. A crosslaminata according to claim 4, characterised in that in each of said films A and B the main layer consists of HDPE or LLDPE or a blend of the two, the second bonding layer mainly consists of LLDPE but with admixture of 5-25% of a copolymer of ethylene having a melting point or a melting range within the temperature interval 50-80 degr. C, and the strands mainly consist of a copolymer of ethylene having a melting point or a melting range within the temperature interval 50-100 degr. C or a blend of such copolymer and LLDPE containing at least 25% of the said copolymer.

10. A crosslaminata according to any of the preceding claims, characterised in that it comprises on one or each of the outer films of the laminate, a surface layer which also is surface layer of the laminate and is adapted to enhance heatsealing of the laminate and/or increase its frictional properties.

11. A method of manufacturing a crosslaminated comprising mutually bonded polymer films of which at least two neighbour films (A and B) each are formed by coextruding in a flat or circular die a layer of a polymer material which is selected for high tensile strength (hereinafter the main layer) and a layer (hereinafter the first bonding layer) from a different polymer material which is selected to produce or enhance the bonding between A and B in the lamination process, and in which A and B each is supplied with a uniaxial or unbalanced biaxial molecular orientation at any stage after the joining of the different materials in the coextrusion die and before the lamination, and prior to the lamination A and B are arranged in such a way that the main direction of orientation in A will cross the main direction of orientation in B, and during the lamination the bonding between A and B is established at least in part through heat, and is made strong in spots or lines but is made weaker or is avoided over the rest of the contacting surfaces, characterised in that in the coextrusion each of the said first bonding layers is made a discontinuous layer consisting of an array of strands and in the lamination the array of strands on A are arranged to cross the array of strands on B, and the heat is applied generally evenly all over A and B and is adapted to make the strands on A strongly bond to the strands on B in the spots where they intersect the latter but make a weaker bonding or avoid bonding over the parts of the contacting surfaces, which are devoid of any first bonding layer.

12. A method according to claim 11, in which the coextrusion of at least one of the films A or B is carried out by means of a circular coextrusion die, to form and draw-down a tubular film characterised in that the draw-down is adapted to produce a significant uniaxial or unbalanced biaxial maltoorientation with the main direction of orientation and the direction of the array of strands either extending along the longitudinal direction of the film or, by means of a relative rotation between the exit of the die and means to take up the film after the extrusion is made to extend helically along the tubular film, and subsequently the film is cut open under an angle to the main direction of orientation and to the direction of the array.

13. A method according to claims 11 and 12, characterised in that the distance from the middle to middle of neighbour strands at the exit from the extruder is at the highest 8 cm, preferably no higher than 4 cm and more preferably no higher than 2 cm, and the circumference of the tube at this exit is at least 20 cm.

14. A method according to any of the claims 11-13, characterised in that in the coextrusion process, A and/or B are also supplied with a continuous surface layer (hereinafter the second bonding layer) which is coextruded on the main layer under the array of strands, whereby the said second bonding layer consists of a polymer material different from those in the main layer and in the first bonding layer, and is selected to produce, during the lamination, bonding also at locations which are devoid of

any first bonding layer, but a bonding of lower strength than the bonding in the spots.

15. A method according to any of the claims 11-14, characterised in that the described arrays of strands are coextruded on both sides of A, and in the lamination process there is applied a B-film on each side of A with the arrays of A on each side intersecting with arrays of a B-film.

16. A method according to claim 14 in which in addition to the films A and B there is applied at least one more film in the lamination, characterised in that said film also is produced by coextrusion and thereby is provided with a surface layer of a composition adapted to control its bonding in the laminate, whereby this composition and the lamination conditions are chosen such that the strength of this bonding becomes higher than the bonding strength between A and B at the locations which are devoid of the coextruded strands.

17. A method according to any of the claims 11-16, characterised in that following the bringing-together of the films in a sandwich arrangement for lamination, before or after the bonding of said sandwich arrangement to a laminate by heat, the films are further oriented by stretching in the longitudinal and/or in the transverse direction.

18. A circular extrusion die comprising a distribution part in which at least a first molten polymer material can be formed into a generally even circular flow, and bodily

separate from this an exit part comprising a circular main channel with generally cylindrical or conical walls, which channel may comprise a flat zone, to conduct said molten polymer material towards an exit orifice from which it will leave the die as a tubular film structure, characterised in that said exit part also comprises a channel system for circumferential extrusion of a circular array of narrow strands of a second molten polymer material, said channel system ending in a circular row of internal orifices in the outward generally cylindrical or conical wall of the main channel.

19. A circular extrusion die according to claim 18, characterised in that said circumferential extrusion starts at one or a few inlets to the exit part and comprises for equal dividing a labyrinthine channel system starting at each inlet, each such system comprising at least three channel-branchings.

20. A circular extrusion die according to claim 19, characterised in that the channels of the labyrinthine system or systems terminate in a common circular channel having a wall common with a part of the generally cylindrical or conical wall of the main channel, the circular row of internal orifices being located in said wallpart.

21. A circular extrusion die according to any of the claims 18-20, characterised in that the circumference of the inward wall at the exit is at least 20 cm, and the distance from middle to middle of neighbour orifices in the circular row

is adapted to produce, after the magnification or reduction which will happen if the walls of the main channel are generally conical, a distance from middle to middle of neighbours of the strands which is at the highest 8 cm, preferably no higher than 4 cm and more preferably no higher than 2 cm.

22. A circular extrusion die according to any of the claims 18 to 21, in which additionally to the means for coextruding the said first and second molten polymer materials there are means for coextruding a circular flow of a third molten polymer material on the side of the first material which is opposite the second material, characterised in that channel arrangements for joining the flows of first and third materials are provided either in the said distribution part, or in a part between the latter and the bodily separate exit part.

23. Any combination of apparatus which is suitable for carrying out the method according to any of the claims 11-17.

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Fig. 1

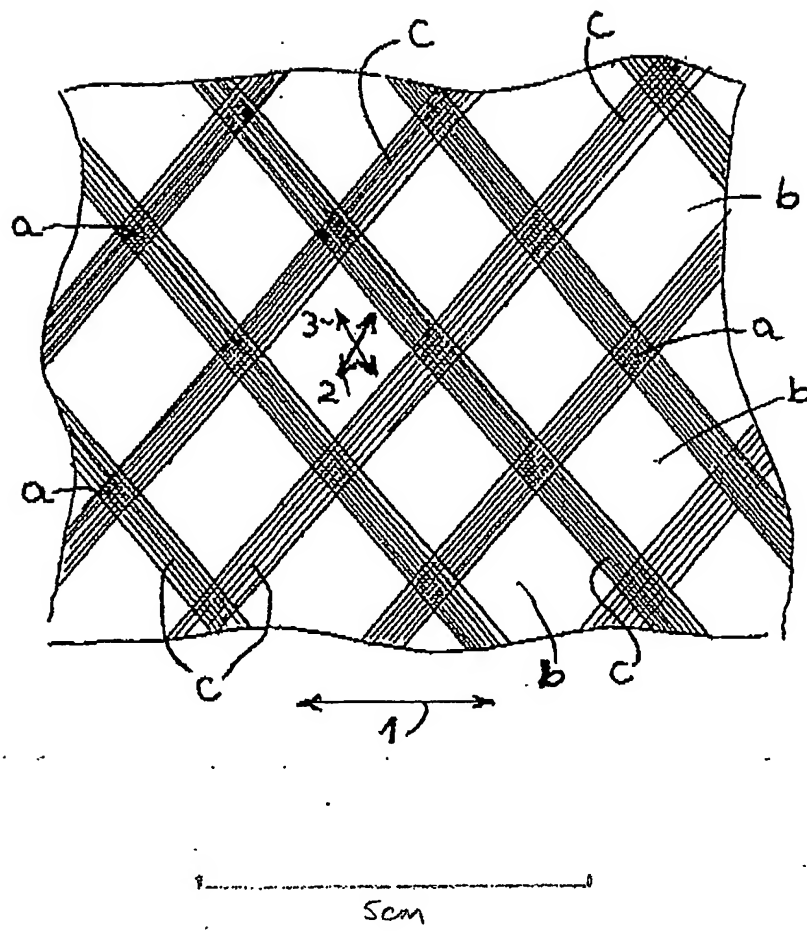


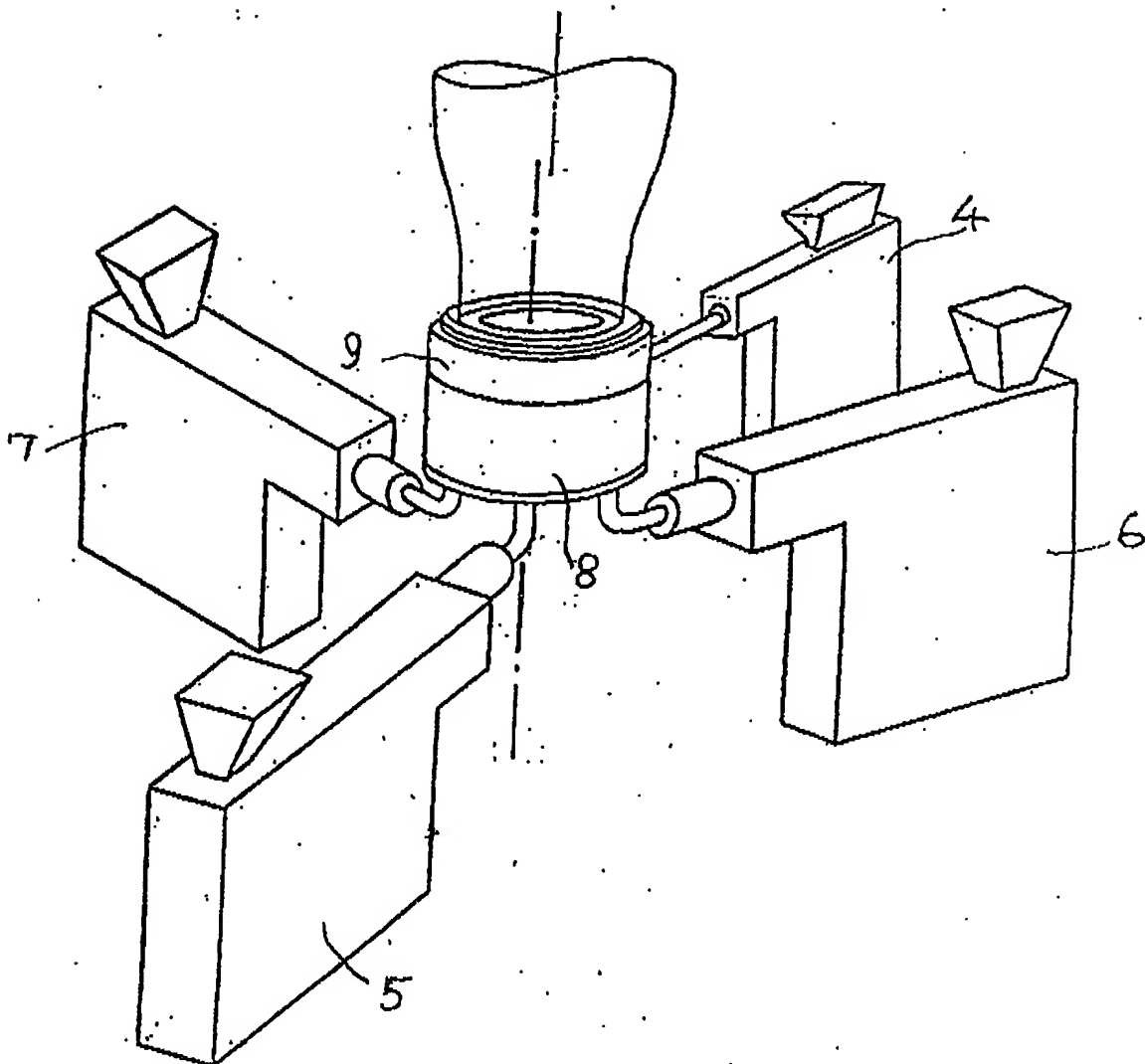
Fig. 2

Fig. 4

